PATENT
TH-2306 02 (US)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF APPEALS AND INTERFERENCES

In re the application of	)	
	)	
PAUL M. McALLISTER ET AL	)	
	)	
Serial No. 10/815,276	)	Group Art Unit: 1797
	)	
Filed April 1, 2004	)	Examiner: Boyer, Randy
	)	
REACTOR SYSTEM AND PROCESS FOR THE MANUFACTURE OF ETHYLENE OXIDE	)	June 11, 2009

COMMISSIONER FOR PATENTS
P. O. Box 1450
Alexandria, VA 22313-1450

Sir:

APPEAL BRIEF

The Appellant hereby files this Appeal Brief in response to the rejection of September 29, 2008, rejecting claims 1-4, 8-14, 19, 21-23, 27-33 and 37-43. The rejection of each of these claims is herewith appealed.

The Commissioner for Patents is hereby authorized to deduct the appropriate fee from Shell Oil Company, Deposit Account No. 19-1800, for filing this Brief on Appeal and for an extension of time under 37 C.F. R. 1.136(a). Appellant respectfully requests that the Examiner's rejections be overturned.

Real Party in Interest

The real party in interest is Shell Oil Company.

Related Proceedings

There are no related proceedings.

Status of Claims

Claims 1-36 were originally presented for examination. In the amendment and response of July 13, 2007, claims 1 through 14 were amended, claims 15 through 18 were canceled, claims 19 through 33 were amended, and claims 34 through 36 were canceled.

In the amendment of February 7, 2008, claims 1 through 3 were amended, claims 5 through 7 were canceled, claims 10 and 11 were amended, claim 19 was amended, claim 20 was canceled, claims 21 and 22 were amended, claims 24 through 26 were canceled, claims 29 and 30 were amended and new claims 37 through 43 were added.

Status of Amendments

No amendments were made subsequent to the final rejection or after the office action of September 29, 2008.

Summary of Claimed Subject Matter

Claim 1 describes a reactor system for the oxidation of ethylene to ethylene oxide (see the specification, page 2, lines 8-13) which comprises an elongated tube having a reaction zone defined by a tube length and a tube diameter (specification, page 2, lines 20-21) wherein the tube diameter is at least 28 millimeters (specification, page 3, lines 2-3). Contained within the reaction zone is a packed bed of shaped support material (specification, page 2, lines 21-23) wherein the shaped support material has a hollow cylinder geometric configuration defined by an inside diameter, an outside diameter and a length (specification, page 2, lines 23-25). The ratio of the length to the outside diameter is from 0.5 to 2 (specification, page 2, lines 30-32) and the ratio of the outside diameter to the inside diameter exceeds 2.7 (specification, page 3, lines 2-4) and the ratio of the tube diameter to the outside diameter is from 2 to 10 (specification, page 3, lines 4-6) and wherein the outside diameter is in the range of 7.4 to 11.6 millimeters (specification, page 21, line 20 through page 22, line 19).

Claim 19 describes a reactor system for the oxidation of ethylene to ethylene oxide (see the specification, page 2, lines 8-13) which comprises an elongated tube having a reaction zone defined by tube length and a tube diameter (specification, page 2, lines 20-21) wherein the tube

diameter is at least 28 millimeters (specification, page 3, lines 2-3). Contained within the reaction zone is a packed bed of shaped support material (specification, page 2, lines 21-23) wherein the shaped support material has a hollow cylinder geometric configuration defined by an inside diameter, an outside diameter and a length (specification, page 2, lines 23-25). The ratio of the length to the outside diameter is from 0.5 to 2 (specification, page 2, lines 30-32) and the ratio of the outside diameter to the inside diameter exceeds 2.7 (specification, page 3, lines 2-4) and the ratio of the tube diameter to the outside diameter is from 2 to 10 (specification, page 3, lines 4-6) and wherein the outside diameter is in the range of 7.4 to 11.6 millimeters (specification, page 21, line 20 through page 22, line 19). The ratio of the outside diameter to the inside diameter provides a positive test result defined by a decrease of the quotient of a numerical value of the pressure drop per unit length of the packed bed and a numerical value of the packing density, which numerical values are obtained by testing the packed bed in a turbulent flow of nitrogen gas at a pressure of 1.136 MPa (specification, page 3, lines 7-24). The support material has a nominal outside diameter of 8 millimeters and a nominal inside diameter of 3.2 millimeters and a ratio of the nominal length to the nominal outside diameter of 1 (specification, page 3, lines 24-33).

Grounds of Rejection to be Reviewed on Appeal

The only ground of rejection to be reviewed upon this appeal is whether or not the appealed claims are obvious under Section 103(a) in view of Tamura (US 4,645,754) combined with Saito (US 4,511,671) or alternatively in view of Tamura (US 4,645,754) combined with Saito (US 4,511,671) and Murphy (US 4,358,623).

Argument

Dr. Paul M. McAllister, the inventor of this application, submitted a Declaration Under Rule 132 (Exhibit A). He describes the inventive reactor system as providing an improved balance of the tube packing density, also the bed voidage and the catalyst hold-up, relative to the pressure drop across the packed bed when in use in ethylene oxide manufacturing, as compared to conventional systems. This is discussed in paragraph 6 of the Declaration.

Dr. McAllister states that Fig. 1 shows the unexpected improvement. The data in Fig. 1 plots the "test result" versus the inside diameter to outside diameter ratio of the cylinders. The "test result" is defined by the quotient of a numerical value of the pressure drop per unit length of the packed bed and a numerical value of the packing density. The numerical values are obtained

as described in the second paragraph of section 6 of the declaration. A decrease in the quotient is considered a positive test result and signifies an improved balance of the tube packing density relative to the pressure drop across the packed bed.

Dr. McAllister states that Fig. 1 shows that the "test result" increases for inside diameter/outside diameter ratios approaching zero and also those greater than 0.37 (which is an outside diameter to inside diameter ratio of less than 2.7). Dr. McAllister states that increases in the test result indicate a less desirable geometery (i.e., the pressure drop penalty is more significant than the packing density benefit or the tube packing density penalty is more significant than the pressure drop benefit).

Dr. McAllister states that this is truly unexpected in light of what would have been predicted for pressure drop by a commonly accepted scientific correlation known as the Ergun Correlation. The correlation is described in detail in the Declaration.

Dr. McAllister describes the experiments which were recorded in Example III of the application. For all of the experiments, the parameters in the Ergun Correlation were calculated from the hollow cylinder characterization information as described in the declaration.

The calculated information (the Ergun Correlation expected test result) based on the predicted pressure drop was compared against the actual test result described in the example. The results are shown in Fig. 2. It can be seen that much higher pressure drop values were predicted by the Ergun Correlation with reduced inside diameter and the test result increased monotonically. <u>In contrast</u>, the experimental results demonstrated a minimum value of the test result (compare the upper line on the graph based on the solid squares to the lower line on the graph based on the unfilled in squares). This demonstrates an unexpected benefit in the balance of pressure drop and packing density with reduced inside diameter cylinders.

Dr. McAllister goes on to state that it was <u>truly</u> <u>unexpected</u> that the combination of a large tube diameter (at least 28 millimeters) and a hollow cylinder geometery for nominal 8, 9, 10 and 11 millimeter supports having a small inside diameter (as is claimed in the present invention) would result in an improved balance of tube packing density relative to the pressure drop across the packed bed. In particular, he stated that it is unexpected that the tube packing density is improved relative to the standard 8 millimeter outside diameter support without the expected increase in pressure drop across the packed bed. It would have been predicted by the

Ergun Correlation that support shapes with such small diameters would not be acceptable for commercial ethylene oxide production.

Tamura relates to the production of ethylene oxide by catalytic vapor phase oxidation of ethylene with oxygen. The invention of Tamura relates to the use of a carrier in the shape of Intalox or Berl saddles in forming the silver-based catalyst. *See U.S. Patent No. 4,645,754*, col. 2, ll. 47-56. Tamura teaches that the saddle shape of the catalyst is an improvement over Raschig rings (i.e., hollow cylinders) in that high selectivity is observed and only minimal pressure loss is observed in the catalyst bed. *See Id.* at col. 3, ll. 29-40. Further, Tamura teaches that the ratio of apparent surface area of the catalyst to the apparent volume is desired to be large. With respect to Raschig rings, Tamura teaches that a decrease in their wall thickness is effective in increasing the ratio of apparent surface area of the catalyst to the apparent volume; however, decreasing the wall thickness is disadvantageous. *See Id.* at col. 4, ll. 13-18. With respect to Raschig rings (which are hollow cylinders), Tamura also states:

> "Further, when the packing specific gravity in a catalyst using a carrier in the shape of spheres or Raschig rings is equalized with that in a catalyst using a porous inorganic refractory carrier in the shape of Intalox saddles or Berl saddles, the former catalyst does not acquire so high selectivity and so low pressure loss as attained by the latter catalyst." *See Id.* at col. 4, ll. 44-51.

Saito relates to the production of <u>methacrolein</u> by catalytic vapor phase oxidation of isobutylene or tertiary butanol with oxygen. *U.S. Patent No. 4,511,671* col. 1, ll. 6-10. Saito teaches to increase the geometrical surface area of the hollow cylinder shaped catalyst to improve conversion performance (i.e., methacrolein formed in the pores diffuses more quickly than in a solid cylinder reducing further reaction of the methacrolein into other compounds such as methacrylic acid, acetic acid, carbon dioxide and carbon monoxide) and improve pressure drop in the catalyst layer <u>compared to a spherical or solid cylindrical shape</u>. *Id.* at col. 1, ll. 35-44; col. 2, ll. 25-50.

The Examiner asserts: "Therefore, the person having ordinary skill in the art of reactor systems for the oxidation of ethylene would have been motivated to use catalyst support materials of the type disclosed by Saito in the process of Tamura in order to achieve higher catalyst activity and higher yield while maintaining a reduced pressure drop across the catalyst bed." *See Office Communication dated April 29, 2008*, page 5, second full paragraph.

As discussed in the declaration by Dr. McAllister (Exhibit A), point number 10, the oxidation reaction of Saito is a diffusion-limited reaction whereas the ethylene epoxidation reaction is not. There are different considerations to be taken into account for a diffusion-limited reaction than for the reaction of the present invention. Therefore, he concluded that the skilled person would not have been motivated to use the catalyst support materials of the shape disclosed in Saito in the ethylene epoxidation process of Tamura in order to achieve higher catalyst activity and higher yield.

In the declaration, Dr. McAllister states that Saito teaches that the disclosed catalyst has improved selectivity and yield compared to <u>spherical or solid cylinder shapes</u>. He states that the key to the improvement in catalyst performance is attributed to the increase in the geometric surface area of the catalyst compared to a <u>solid</u> <u>cylinder</u>. The increase in geometric surface area allows the methacrolein product to diffuse more rapidly than with a solid cylinder, thus reducing the consecutive reaction of methacrolein to methacrylic acid, acetic acid, carbon dioxide and carbon monoxide (he cites the patent at column 1, lines 35-44; column 1, line 51-column 2, line 20; column 2, lines 25-3). Based on this, Dr. McAllister concludes that the methacrolein reaction of Saito et al is a <u>diffusion-limited reaction with an effectiveness factor significantly less than one</u>.

Dr. McAllister states that intrapellet diffusion is correlated by an effectiveness factor (i.e., actual reaction rate/reaction rate ratio without the influence of pore diffusion). The maximum value for the effectiveness factor is one.

Dr. McAllister states that the ethylene epoxidation system <u>is</u> <u>not</u> a diffusion-limited reaction and therefore has an effectiveness factor of essentially one for the supports described in the present application.

Dr. McAllister states that in a diffusion-limited reaction system, as the diameter of a pellet is reduced, the effectiveness factor increases. Also, for a hollow cylinder, effectiveness factor increases as the inside diameter is <u>increased</u> relative to the outside diameter. He states that these trends are observed in the working examples of Saito et al. and that this reinforces that it is a diffusion-limited reaction.

Dr. McAllister describes how Fig. 3 in Section 10 of the declaration demonstrates how differently the behavior of Saito et al. system is compared to the ethylene epoxidation system. The relative yields of the two different types of reactions were plotted versus the relative density

for six millimeter outside diameter cylinder data of Examples 1-3 and Comparative Examples 1-2 of Saito et al. The relative yield was also plotted for a higher selectivity catalyst for ethylene epoxidation. The ethylene epoxidation data for several different relative density values was obtained using a predictive model as described in the declaration.

Dr. McAllister states that based on the results from Saito et al., the yield of the desired product increases with lower mass of catalyst in the reactor. However, for the ethylene epoxidation system, the yield clearly benefits from greater catalyst mass. Based on this, Dr. McAllister concludes that Saito et al. is a diffusion-limited reaction whereas the ethylene epoxidation reaction is not. He goes on to state that there are different considerations that will be taken into account for each of these different types of reactions. In particular, for the diffusion-limited system in Saito et al., one skilled in the art would want to increase the effectiveness factor by reducing the pellet size and/or increasing the inside diameter to decrease the mass of the catalyst in the reactor. However, in the present invention, the inside diameter is decreased for large outside diameter supports in order to improve the tube packing density (i.e. greater mass in the reactor) which also unexpectedly improves the balance of tube packing density relative to the pressure drop across the packed bed.

The Appellants assert that this makes is clear that one of ordinary skill in the art would not have thought to combine the disclosure of Saito et al. with the disclosure of Tamura to improve the pressure drop in the reactor tubes.

In response to this argument, the Examiner states in paragraph 26 on page 12 of the Office Action of September 29, 2008, that Tamura discloses cylindrical shaped support materials for use in its reaction. However, this is not responsive because the Examiner's rejection is based on combining the specific geometry and size limitations of the support materials described in Saito with the disclosure of Tamura. Clearly, because they are different types of reaction systems, one would not be lead to combine the specific geometry and size limitations of Saito with the disclosure of Tamura.

Saito does disclose that there is a decrease in pressure drop for hollow cylinder or ring-like shaped catalysts, but this is **in comparison to solid spheres or solid cylinders**. *See U.S. 4,511,671*, col. 1, ll. 39-44; col. 2, ll. 34-50. The skilled person would not have been motivated to use the catalyst support materials of the type disclosed in Saito in the ethylene epoxidation process of Tamura in order to maintain a reduced pressure drop across the catalyst bed since the

improved pressure drop discussed in Saito is in comparison to solid spheres or cylinders and not the saddles of Tamura. Tamura clearly discloses that Intalox or Berl saddles are considered superior to Raschig rings (i.e., hollow cylinders) and thus tends to teach away from the claimed invention.

The Examiner asserts that Saito uses a 25.4 mm reaction tube in the examples and contemplates use of his system for industrial production which implies a scale-up to a larger tube size. Dr. McAlister states in the Declaration in Paragraph 11 that the tube diameter of 25.4 mm <u>is of industrial scale</u> and that one of ordinary skill in would scale-up Saito's system by multiplication (use a multitubular reactor) rather than increase the tube diameter because one of the objects of Saito is to eliminate hot spots in the catalyst layer in the reactor.

Murphy discloses at column 3, lines 50 through 53 that "packing the tubes with small solid bodies such as beads can result in a substantial obstruction to gas flow with resulting increase in pressure drop." Murphy also discloses at column 4, lines 32 through 35 that "pressure drop through the tubes containing the balls is significantly less than that experienced with tubes packed with beads (i.e., balls of a very small size)." As discussed in the declaration by Dr. McAllister, point number 12, this disclosure in Murphy is in agreement with the Ergun Correlation which would predict an increase in pressure drop per unit length for beads compared to larger balls. Thus, Murphy adds nothing that is helpful to the Examiner's position.

<u>Conclusion</u>

Thus, claim 1 is non-obvious over Tamura in view of Saito, or alternatively over Tamura in view of Saito as evidenced by Murphy. The arguments above also apply to claim 19; therefore, claim 19 is also non-obvious over Tamura in view of Saito, or alternatively over Tamura in view of Saito as evidenced by Murphy. The remaining claims depend from claims 1 or 19; therefore, they are also non-obvious over over Tamura in view of Saito, or alternatively over Tamura in view of Saito as evidenced by Murphy. Appellant respectfully requests the rejection be overturned.

Respectfully submitted,
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CLAIMS APPENDIX

1. A reactor system for the oxidation of ethylene to ethylene oxide comprising:

an elongated tube having a reaction zoned defined by a tube length and a tube diameter, the tube diameter being at least 28 mm; wherein contained within the reaction zone is a packed bed of shaped support material; and wherein the shaped support material has a hollow cylinder geometric configuration defined by a length an outside diameter and an inside diameter such that the ratio of the length to the outside diameter is in the range of from about 0.5 to about 2, and further such the ratio of the outside diameter to the inside diameter exceeds about 2.7, and the ratio of the tube diameter to the outside diameter is in the range of from about 2 to about 10; wherein the outside diameter is in the range of from about 7.4 mm to about 11.6 mm.

2. The reactor system as recited in claim 1, wherein

the tube diameter is in the range of from 28 mm to about 60 mm, and

the ratio of the outside diameter to the inside diameter is

in the range of from about 3 to about 23.

3. The reactor system as recited in claim 1, wherein the tube diameter is in the range of from 28 mm to about 60 mm, and

the ratio of the outside diameter to the inside diameter is in the range of from about 3.3 to about 10.

4. The reactor system as recited in claim 1, wherein the tube diameter is about 39 mm.

5. (Canceled).

6. (Canceled).

7. (Canceled).

8. The reactor system as recited in claim 1, wherein the tube length is in the range of from about 3 to about 15 meters.

9. The reactor system as recited in claim 1, wherein at least 50 percent of the packed bed comprises the shaped support material.

10. The reactor system as recited in claim 1, wherein the ratio of the tube diameter to the outside diameter is in the range of from about 2.5 to about 7.5.

11. The reactor system as recited in claim 10, wherein the ratio of the tube diameter to the outside diameter is in the range of from about 3 to about 5.

12. The reactor system as recited in claim 1, wherein the shaped support material comprises predominantly alpha-alumina, and the packed bed has a tube packing density greater than about 550 kg per cubic meter.

13. The reactor system as recited in claim 1, wherein the shaped support material supports a catalytic component.

14. The reactor system as recited in claim 13, wherein the catalytic component comprises silver.

15-18. (Canceled)

19. A reactor system for the oxidation of ethylene to ethylene oxide comprising:

an elongated tube having a reaction zone defined by a tube length and a tube diameter, the tube diameter being at least 28 mm; wherein contained within the reaction zone is a packed bed of shaped support material; and wherein the shaped support material has a hollow cylinder geometric configuration defined by a length, an outside diameter and an inside diameter such that

the ratio of the length to the outside diameter is in the range of from about 0.5 to about 2, and

the ratio of the outside diameter to the inside diameter provides a positive test result, and further such that

the ratio of the tube diameter to the outside diameter is in the range of from about 2 to about 10;

wherein the ratio of the outside diameter to the inside diameter exceeds about 2.7;

wherein the outside diameter is in the range of from about 7.4 mm to about 11.6 mm ; and

wherein the "positive test result" is defined by a decrease of the quotient of a numerical value of the pressure drop per unit length of the packed bed and a numerical value of the packing density, which numerical values are obtained by testing the packed bed in a turbulent flow of nitrogen gas at a pressure of 1.136 MPa (150 psig), relative to a comparison quotient of

numerical values obtained in an identical manner, except that the hollow cylinder geometric configuration of the same support material is defined by

a nominal outside diameter of 8 mm and a nominal inside diameter of 3.2 mm, and

a ratio of the nominal length to the nominal outside diameter of 1.

20. (Canceled).

21. The reactor system as recited in claim 19, wherein

the tube diameter is in the range of from 28 mm to about 60 mm, and

the ratio of the outside diameter to the inside diameter is in the range of from about 3 to about 23.

22. The reactor system as recited in claim 19, wherein the tube diameter is in the range of from 28 mm to about 60 mm, and

the ratio of the outside diameter to the inside diameter is in the range of from about 3.3 to about 10.

23. The reactor system as recited in claim 19, wherein the tube diameter is about 39 mm.

24. (Canceled).

25. (Canceled).

26. (Canceled).

27. The reactor system as recited in claim 19, wherein the tube length is in the range of from about 3 to about 15 meters.

28. The reactor system as recited in claim 19, wherein at least 50 percent of the packed bed comprises the shaped support material.

29. The reactor system as recited in claim 19, wherein the ratio of the tube diameter to the outside diameter in the range of from about 2.5 to about 7.5.

30. The reactor system as recited in claim 29, wherein the ratio of the tube diameter to the outside diameter in the range of from about 3 to about 5.

31. (Previously Presented) The reactor system as recited in claim 19, wherein the shaped support material comprises predominantly alpha-alumina, and the packed bed has a tube packing density greater than about 550 kg per cubic meter.

32. The reactor system as recited in claim 19, wherein the shaped support material supports a catalytic component.

33. The reactor system as recited in claim 32, wherein the catalytic component comprises silver.

34-36. (Canceled)

37. The reactor system as recited in claim 1, wherein the outside diameter is in the range of from about 7.4 to about 8.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 3 to about 15.

38. The reactor system as recited in claim 1, wherein the outside diameter is in the range of from about 8.4 to about 9.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 3 to about 15.

39. The reactor system as recited in claim 1, wherein the outside diameter is in the range of from about 9.4 to about 10.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 4 to about 10.

40. The reactor system as recited in claim 1, wherein the outside diameter is in the range of from about 10.4 to about 11.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 4.6 to about 10.

41. The reactor system as recited in claim 19, wherein the outside diameter is in the range of from about 7.4 to about 8.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 3 to about 15.

42. The reactor system as recited in claim 19, wherein the outside diameter is in the range of from about 8.4 to about 9.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 3 to about 15.

43. The reactor system as recited in claim 19, wherein the outside diameter is in the range of from about 9.4 to about 10.6 mm and the ratio of the outside diameter to the inside diameter is in the range of from about 4 to about 10.

EVIDENCE APPENDIX

Exhibit A: Declaration Under Rule 132 of Dr. Paul M. McAllister.

RELATED PROCEEDINGS APPENDIX

There are no related proceedings.